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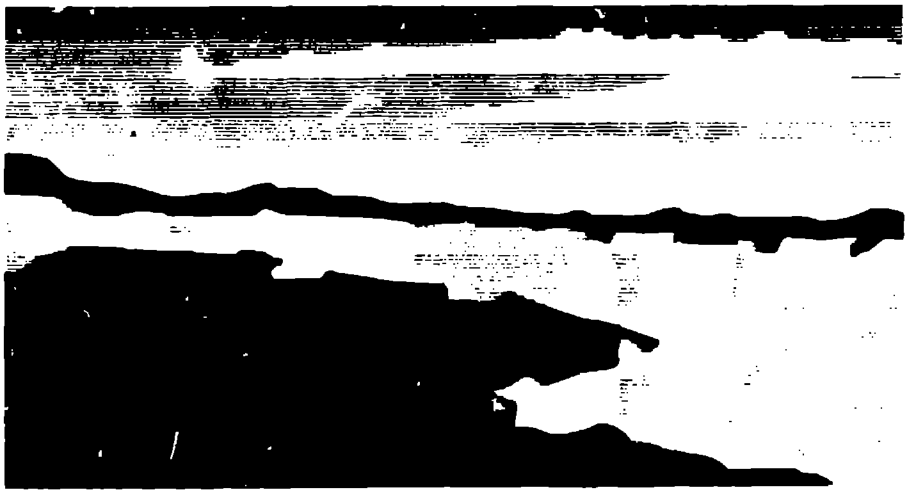

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Behavior of Structural and Target Materials Irradiated in Spallation Neutron Environments

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Abstract. This paper describes considerations for selection of structural and target materials for accelerator-driven neutron sources. Due to the operating constraints of proposed accelerator-driven neutron sources, the criteria for selection are different than those commonly applied to fission and fusion systems. Established irradiation performance of various alloy systems is taken into account in the selection criteria. Nevertheless, only limited materials performance data are available which specifically related to neutron energy spectra anticipated for spallation sources.

INTRODUCTION

The development of accelerator-driven neutron sources provides advantages over fission and fusion neutron sources in terms of source accessibility and control, but presents some unique challenges regarding the selection and use of structural and target materials. Many advantages of proposed systems lie in the fact that the neutron production is confined to the target material or the target material surrounded by a neutron multiplier. In some configurations, for instance the power-producing accelerator transmutation of waste system, a fission-based multiplying medium surrounds the target region to produces additional neutrons in a fission blanket. However, even with the fission source present, the system is subcritical and is easily shut down by stopping the accelerator-driven neutron supply.

Because of these attributes, accelerator-driven neutron systems are much different than present fission reactor systems and proposed fusion reactor systems. In the case of fission reactors, the most widely utilized designs, light water reactors, operate at high pressures (from 1000 to 2200 psi depending on the design) and moderate temperatures (~300°C) for metallic (i.e. non-fuel) components. These operating constraints, which are set by neutron moderation and heat removal requirements, mean that those systems require large, thick section pressure vessels, which in turn restricts materials selection to medium yield strength steels with high fracture toughnesses. The principal safety issue for those systems is related to the fracture resistance of the pressure vessel, which is known to degrade with neutron irradiation.

Fusion systems designs are still not yet well enough established to uniquely quantify materials operating constraints. However, major attention is being directed to materials capable of handling high intermittent heat fluxes. The first wall of the fusion chamber must not only provide a hard vacuum seal, but also be able to conduct away large heat impulses during plasma burn periods, and withstand large numbers of the associated thermal-mechanical load cycles.

Accelerator-driven systems tend not to be restricted by high pressure and in many cases high temperature, except local to the target material, which serves only minor structural capacity. This opens the possibility of selecting from a much wider group of materials for structural service because of the relatively less stringent requirements regarding potential fracture and high temperature applications. Nevertheless, other structural material selection restriction apply due to the unique nature of the operating environments in proposed accelerator-driven neutron sources. These restriction will be discussed here in light of the understanding of materials performance in similar spallation neutron environments, and in fission neutron and other spectra. The relevant materials database is small, but some generalizations can be drawn from existing information, and plans for the development of experimental programs to start to acquire other relevant information can be drawn.

PROBLEM DEFINITION

Materials selection for accelerator-driven neutron sources is highly dependent on the influence of the irradiation field on materials properties. It is, therefore, necessary to examine a number of open issues which affect this performance, and for which significant experimental programs have been developed to address these issues in fission and fusion applications. The major issues affecting irradiation stability can be listed as follows: 1. the initial atom displacement process, 2. the efficiency of the damage production - surviving defects and their configuration, 3. the agglomeration of defects into defect clusters, and 4. defect cluster evolution with fluence, and 5. the influence of the numbers and nature of the defect clusters on physical and mechanical properties of the material. Due to the differences of the irradiating and secondary particle energies from high energy proton-induced spallation sources, it is not immediately clear to what extent present understanding of these issues in fission and fusion applications can be extrapolated to the spallation case. In any case, other materials application issues, such as strength, fracture resistance, corrosion resistance, etc. must also be accounted for in the selection process. This helps to define the number of potential materials systems by virtue of their use and applicability in other similar situations.

IRRADIATION DAMAGE AND TRANSMUTATION ISSUES

The field of irradiation damage has received much attention over the past 40 years, and has resulted in a much better understanding of the role of point defect (e.g. interstitials, vacancies, transmutants) production, migration and agglomeration on materials physical properties and mechanical performance. A companion paper in this conference deals directly with the calculation of the atomic displacement crossections and their use for establishing the initial numbers of defects produced given the neutron or proton energy spectrum [1]. Once the initial defect numbers are established, it is necessary to determine how many survive the initial quenching phase and in what form. This is critical issue for comparison of radiation damage between fission, fusion, and spallation spectra. Again, much effort has been expended to address this issue, particularly with regard to the fission-fusion correlation where a large part of the fusion materials irradiation performance database has been developed in fast or mixed spectra fission reactors. Three basic approaches have been used to try to bracket the problem: computer simulation calculations of displacement cascades and annealing effects, direct observation of defect clusters by transmission electron microscopy, and comparisons of physical or mechanical properties changes in various spectra (i.e. properties-properties correlations). The computer calculations were first to indicate the potential for direct comparison of damage structures in cascades due to highly energetic incident particles [2]. This is because, above a certain incident particle energy, the cascades 'break-up" to form sub-cascades, where the sub-cascade size is nearly constant.

Thus higher energy incident particles form larger numbers of subcascades, but the subcascades which do form are directly comparable to those formed through lower energy knock-ons as long as the energies are above the cut-off. Direct observation of this effect has been made in certain materials [3,4], and has also been confirmed by properties-properties correlations in other systems [5]. This comparability of cascade structure bodes well for allowing useful materials performance data and trends to be obtained from the large body of fission irradiations.

One major difference between spallation-based sources and fission sources is the differences in the amounts and types of transmutation produces. Many transmutation reaction require a threshold neutron energy which is on the order of 2 to 10 MeV, usually above most fission neutron energies, but well within the range of neutron produced in DT fusion or by spallation. Particular concerns have been expressed regarding the amounts of H and He produced in materials systems under irradiation with highly energetic incident particles. A good deal of effort has been put forward to assess the extent of this problem for fusion applications. Presently opinions of the effect vary since it is postulated that insoluble inert gasses, particularly He, will stabilized void formation, and possibly contribute to high temperature embrittlement by collection along grain boundaries. Evidence to support this picture has been presented, but the extent of the effect seems not to be as damaging as first postulated. The production of these and other types of transmutants in spallation sources has been investigated [6].

Based on the foregoing, it should be evident that previous materials investigations and application experience for neutron environment uses provides a insight for spallation source application, with caution.

MATERIALS PERFORMANCE AND SELECTION CRITERIA

Materials performance can be generalized by considering various ranges of temperature over which properties apply. Table 1 gives approximate characterizations of issues of selection which are of concern over various temperature ranges. Since several of the properties depend on thermal activation and the mobility (or lack thereof) of point defects, the generalization can be very useful for identifying properties which control materials performance.

Table 1. Temperature Regimes of Interest and Associated Materials Performance Concerns

$T < 0.3\ T_m$ (T_m is the melting temperature)

- No or low swelling
- Embrittlement (Ductility)
- Hardening (Strength)
- Thermal creep not a concern, transient irradiation creep may occur
- H embrittlement may be important
- He effects may be important
- Irradiation-induced precipitation, segregation minimal
- Corrosion minor concern
- Fatigue and Crack Growth

$0.3\ T_m < T < 0.55\ T_m$

- Swelling
- Creep and Irradiation Creep
- Hardening and Embrittlement (Ductility)

H effects less than at low temperature, H attack
He effects are important
Irradiation-induced precipitation and segregation are important
Corrosion important
Fatigue and Crack Growth

$T > 0.55\ T_m$

Smaller swelling, but rate effects and He effects may contribute
Hardening less concern
Embrittlement by radiation-induced segregation and He
Low mechanical strength
Thermal creep
High corrosion rates

Based on several years of research, development and application of a variety of materials systems for neutron environments, several classes of alloys are worthy of consideration for spallation environments. These are listed in Table 2.

Table 2, Classes of Materials for Examination

Structural Materials
- Aluminum and Alloys
- Ferritic and Martensitic Steels (Fe and Fe-Cr base)
- Stainless Steels and High Alloy Fe-Ni-Cr Materials
- Zirconium and Alloys
- Vanadium and Alloys
- Titanium and Alloys

Target Materials
- Pb-Bi, Pb (liquid or solid)
- High Z refractories and Alloys (W)
- U and Alloys
- Others, Th

Barrier and Coating Materials
- Ceramics
- Coatings/Cladding

Advanced Applications
- Composites for enhance radiation performance and NDE: Al/SiC, SiC/SiC, C/C

The selection and use of barrier and coating materials, and the potential for use of advanced materials systems including metal-matrix and ceramic-matrix composites will not be considered directly here. However, based on the time scale of the development of several of the proposed accelerator-driven technologies, advance materials systems should be available for use at the time of demo-scale plant construction.

IRRADIATION PERFORMANCE OF POTENTIAL STRUCTURAL MATERIALS

The irradiation performance of several of the structural materials are now presented with reference to their durability and limitations by virtue of use or experimental findings in neutron irradiation environments. As will be clear, much of the characterization of performance is based on irradiations in a fission reactor neutron spectra, but limited data are also available for exposure in spallation neutron environments. The spallation neutron irradiation experience is reviewed in another paper in detail [7]. Attention will be directed here a more general overview of the performance of certain classes of materials in neutron environments.

Aluminum and Alloys

Aluminum and it alloys have been of interest for nuclear applications since early fission reactor designs. This material is low Z and has minimal activation problems. High strength structural alloys are available, and are used extensively in applications where high temperature is not a concern. The low melting point of Al and its alloys is the one significant drawback to its use in energy conversion systems where strength at moderately high temperature is important.

Most of the relevant high fluence radiation experience at 55°C with aluminum alloy 6061 (T6 heat treatment) is available from the HFBR at Brookhaven reactor. Al6061 is precipitation strengthened by Mg_2Si. The BNL experience parallels other experimental irradiation work on that alloy. The principal finding of those studies is that the alloy continues to strengthen but loose ductility under irradiation until dose of around 10^{23} n/cm^{2}s are reached when the strengthening and ductility level out. The mechanical properties changes are associated with the thermal neutron transmutation reaction of Al → Si. The additional Mg reacts with the Si to form further precipitates.

The main concern regarding the use of precipitation strengthened aluminum alloys under irradiation is the unresolved question regarding the stability of Mg_2Si (or other) precipitates during neutron versus proton irradiation. One possibility is that the dissolution may be uniquely tied to the radiation-induced cascade structure where the mix of cascade size and number of freely migrating defects affect the precipitate stability. The question of precipitate stability under irradiation requires further consideration.

Table 3. Aluminum Alloy Irradiation Performance

Alloy	Irradiation	Comment	Ref.
6061	n, HFBR-BNL	Thermal n: Al-Si; plateau in prop. changes	[8]
6061	n, HFIR	Thermal n: Al-Si	[9]
5154	n, Petten	GB Mg_2Si embrittlement	[10]
6061	p,n, LAMPF	Mg_2Si dissolution; decrease in strength	[11]
6061, 5054	p,n, LAMPF	Results similar to BNL and ORNL	[12]
6061	He-ions	(Work not completed)	[13]
Others			

Leading Factors:

- Limited to low temperature applications (T_m = 660°C)
- Influence of thermal neutrons on Al → Si conversion
- Influence of high energy neutron/proton irradiation, particularly on precipitate stability

Ferritic and Martensitic Steels

Certain ferritic and martensitic steels, HT-9 in particular, have been shown to be highly radiation damage resistant. Some of this resistance has been explained to be due to intrinsic differences in the properties of body centered cubic (bcc) versus face centered cubic (fcc) alloys. The ferritic martensitic alloy HT-9 has been shown to be high resistant to swelling and to have minimal changes in ductile brittle transition temperature (DBTT) after extended irradiations (> 100 dpa) at temperatures at and above 400°C. However, at temperature around 55°C, the same alloy has been shown to experience substantial shifts in DBTT due to radiation hardening [14,15].

Table 4. Selected Fe-Cr Alloys Irradiation Performance

Alloy	Irradiation	Comment
HT-9	Extensive	Low swelling at 400°C to very high doses (>200 dpa)
9Cr-1Mo	Extensive	Similar to, but worse than HT-9
Other	Extensive	Many other Fe-Cr alloys have been investigated Low activation (for fusion) versions are being developed

Leading Factors:
- Low temperature performance
- Ductile - Brittle transition

Stainless Steels and High Alloy Fe-Ni-Cr Materials

A very large data base exists for austenitic stainless steels in fission reactor applications. These data are primarily for fast fission and fusion reactors with hard neutron spectra. Reactors with a large thermal flux are problematic for high Ni alloys due to a large absorption crossection, and the two step $^{58}Ni(n,\gamma)^{59}Ni$, $^{59}Ni(n, \alpha)^{56}Fe$ reaction which can produce copious amounts of He in large thermal neutron spectra with all of the accompanying problems due to He. Furthermore, activation of these alloys tends to be higher than other acceptable classes of alloys. Because of this experience, austenitic stainless steels and other high nickel Fe-Ni-Cr alloys are often downplayed for nuclear application. However, because of the familiarity with their use, and their relative ease of fabricability and joining, the often end up as the material of choice.

It should be mentioned that certain Fe-Ni-Cr base alloys have been of use in certain spectra anticipated for the ADTT concepts. In particular, Inconel 718 has been used for long periods without problem as a beam window material at LAMPF. Inconel 718 is alsc use in LWR fuel elements, that is high thermal neutron fluxes, for spring materials in the fuel pin plenum region.

Table 5. Irradiation Performance Selected Fe-Ni-Cr Alloy

Alloy	Irradiation	Comment
316 SS	Extensive	Swells a 1%/dpa between 375 and 525°C, after about 20 dpa incubation dose
304 SS	Extensive	similar to 316SS
Inconel 718	LAMPF, LWRs, others	
Other	Extensive	Many other Fe-Ni-Cr alloys have been investigated.

The incubation dose for swelling increases with Ni content. Swelling is highly sensitive to small amounts of alloying element additions.

Leading Factors:

- High He production in thermal neutron fluxes
- High swelling at high doses
- Radiation-induced segregation influences on aqueous corrosion and stress corrosion cracking

Zirconium and Alloys

Zirconium and its alloys have a long standing history of use in water cooled and moderated thermal fission reactor technology. The principal use is as fuel cladding in LWR applications, and for pressure tube and other structural applications in HWRs. The performance of these alloys is well documented. Some typical compositions and applications are listed below.

Table 6. Selected Zirconium Alloy Irradiation Performance

Alloy	Application	Comment
Zircaloy 2	BWR Cladding	Extensive Use
Zircaloy 4	PWR Cladding, IPNS U-clad	Extensive Use
Zr-2.5 Nb	CANDU Pressure Tube Material	Extensive Use
Zirlo	Zr-1Nb-1Sn-.1Fe, PWR Cladding	Now In Use
Zr-1Nb	Cladding in Eastern Block Countries	
Ohzennite	Cladding in Eastern Block untries	

Problems with zirconium and alloys have been established from extensive fabrication and processing histories. Principal problems and solutions are shown below. Comparisons of mechanical properties and corrosion performance have been made [16].

Table 7. Zirconium Performance Problems and Solutions

	Problem	Solution
1.	Hydride Embrittlement	Control H content and texture in fabrication Assure low H uptake in service (improves with Nb content)
2.	Radiation Growth	Controlled by heat treatment during processing
3.	I-induced SCC	Stress corrosion cracking (SCC) can be induced by interaction with fission product I. Th process is sensitive to the precipitate structure in the alloy which can be controlled through initial processing
4.	Loss of ductility	Radiation-induced strengthening and concomitant loss of ductility is common and is taken into account in cladding design life calculations. Zirlo composition my less sensitive to this problem than the others.
5.	High T Reactions	TMI-2 showed that Zr reactions with H_2O and UO_2 quickly produced H_2 and other unwanted products

that lead to severe accident reactor safety issues. This is not an issue for ADTT systems.

Corrosion compatibility with aqueous solutions is adequate. Relatively poor thermal conductivity of this material should not be a concern for ADTT applications, except perhaps for basket materials in a solid pellet target configuration.

Vanadium and Alloys

Vanadium and several of its alloys are of high interest for fusion applications due to the low activation issues. However, vanadium has a very large thermal transmutation crossection V → Cr which renders V unsuitable for mixed spectra reactors. Some of the ADTT applications, particularly the ATW concept, rely on a highly thermalized neutron spectrum. In those cases, the use of V-based alloys may be precluded due to the transmutation problems. Furthermore, swelling in V alloys and the increase of ductile to brittle transition temperature have been shown to be highly sensitive to alloying content. In particular, undersized solutes present severe irradiation effects problems. Present V alloy development has focused on a composition near V-4Ti-4Cr which seems to show good swelling resistance, and only small increases of DBTT with irradiation. There are other limitations for the large scale use of this alloy since there is little or no experience with the production of large forgings or castings of this material. Efforts are underway in the fusion program to address this issue.

Table 8. Irradiation Performance of Vanadium Alloys

Alloy	Irradiation	Comment
V-4Ti-4Cr	Fast Reactor	Lead candidate composition for fusion applications
Several	Fast Reactor	Can control swelling and embrittlement by alloying additions

Leading Factors:

- Large thermal crossection for V → Cr
- High sensitivity to swelling and segregation with undersized solutes
- Ductile - Brittle Transition, Low Fracture Toughness

Titanium Alloys

Titanium alloys have not been the focus of a large research effort for neutron environment application. Thus, the understanding of the performance of this alloys system in neutron environments is limited. Nevertheless, this class of materials has made a significant contribution to the aerospace and aircraft industry where structural demands are also extreme and may have potential for certain applications in ADTT systems. [17]

TARGET MATERIALS SELECTION

There are several outstanding issues for potential target materials, particularly if solid targets are adopted. Table 9 list potential materials and selection considerations.

Table 9. Target Materials

Candidates:
- Pb-Bi, Pb (liquid or solid)
- High Z refractories and Alloys (W)
- U and Alloys
- Others, Th

Issues:
- Liquid Metal:
  - Impurity (transmutant) element effects
  - No structural damage due to irradiation
- Solid:
  - High dpa/s, transmutations/s,
  - Embrittlement
  - Coating/cladding for compatibility
  - Energy Deposition and thermal/mechanical cycles
  - Dimensional stability (U)
  - Transmutation/Activation effects

The issue of target material selection is complex particularly due to the requirements for efficient neutron production and, for solid targets, heat removal considerations. These issues are beyond the scope of the present paper, and are presently being addressed in on-going research work.

SUMMARY

This paper has presented an overview of several aspects of materials selection and potential candidate materials classes primarily for structural application in accelerator-driven transmutation technologies On one hand, it is helpful that similarities and guidance can be drawn from the now vast irradiation damage and materials selection experience for fission and fusion systems. On the other hand, the fission and fusion experience has shown that materials selection is complex, and must be accounted for early in the design and development process. Due to the complexity of the conditions and constraints for use, sufficient data for materials performance assessment is often lacking. Focused experimental programs also need to be directed at the prototypic materials performance characterization early in the design and development process.

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